

What is claimed is:

1. A crystalline form C of entacapone, characterized by the following XRD data:

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Angle 2 theta (°)	Lattice spacing d (Å)	Rel. intensity I/Imax (%)
5.61	15.77	100
11.43	7.78	1
14.75	6.06	2
17.23	5.21	5
18.81	4.78	2
20.89	4.32	1
23.13	3.92	17
25.23	3.62	2
26.87	3.41	3
29.03	3.18	1
32.17	2.90	2

2. A crystalline form D of entacapone, characterized by the following XRD data:

Angle 2 theta (°)	Lattice spacing d (Å)	Rel. intensity I/Imax (%)
6.84	12.95	99
11.84	7.51	6
12.12	7.34	7
13.52	6.59	49
14.8	6.04	23
15.56	5.75	40
16.54	5.42	31
16.9	5.30	22
17.98	4.99	37
18.84	4.77	12
19.06	4.72	13
20.72	4.36	18
21.44	4.22	28
22.24	4.07	12

23.4	3.88	22
24	3.79	39
24.62	3.70	76
25.34	3.60	51
26.5	3.46	65
27.44	3.35	100
28.08	3.28	51
29.24	3.16	15
29.98	3.09	17

3. A crystalline form E of entacapone, characterized by the following XRD data:

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Angle 2 theta (°)	Lattice spacing d (Å)	Rel. intensity I/Imax (%)
6.62	13.35	100
8.87	9.97	4
12.36	7.16	8
12.90	6.86	12
13.38	6.62	11
14.40	6.15	5
15.52	5.71	49
17.92	4.95	33
18.25	4.86	22
19.20	4.62	6
20.48	4.24	26
21.10	4.21	7
21.85	4.07	6
22.45	3.96	6
22.90	3.88	7
24.00	3.71	30
24.64	3.61	36
25.85	3.45	77
27.32	3.26	20

4. A process for the preparation of the crystalline form C of entacapone as claimed in claim 1,

characterized in that entacapone is crystallized from a mixture of at least one aromatic and at least one aliphatic hydrocarbon.

5 5. The process as claimed in claim 4, characterized in that the aromatic hydrocarbon used is toluene and the aliphatic hydrocarbon used is n-heptane.

6. A process for the preparation of the crystalline
10 form D of entacapone as claimed in claim 2, characterized in that

a) entacapone is dissolved in a water-miscible solvent and this solution is added to water or a mixed aqueous system; or

15 b) entacapone is crystallized from a non-acidic solvent or a solvent mixture with at least one non-acidic component, in the presence of a strong acid.

7. The process as claimed in claim 6, variant a),
20 characterized in that it is carried out in THF/water, acetone/water, acetone/DMSO/water or n-propanol/water.

8. The process as claimed in claim 6, variant b),
characterized in that it is carried out in toluene/
25 acetonitrile or toluene/acetonitrile/acetic acid.

9. The process as claimed in claim 6, variant b), or
claim 8, characterized in that the acid used is
hydrogen bromide.

30 10. A process for the preparation of the crystalline form E of entacapone as claimed in claim 3, characterized in that entacapone is dissolved in a polar aprotic or alcoholic solvent and this solution is
35 added to an aliphatic hydrocarbon immiscible with this solvent, in which entacapone is insoluble.

11. The process as claimed in claim 10, characterized in that it is carried out in THF/n-hexane, THF/n-pentane, THF/cyclohexane or isopropanol/n-hexane.

5 12. The process as claimed in one of claims 6-11, characterized in that crude entacapone is used.

10 13. The process as claimed in one of claims 6, variant b), 8 and 9, characterized in that entacapone is used in situ in the form of the product of a Knoevenagel condensation of 3,4-dihydroxy-5-nitrobenzaldehyde and 2-cyanoacetic acid diethylamide.

15 14. The process as claimed in one of claims 6, variant b), 8, 9, 12 and 13, characterized in that the acid used is hydrogen bromide and the process is carried out in toluene/acetonitrile/acetic acid.

20 15. The crystalline form C, D or E of entacapone as claimed in claim 1, 2 or 3 for use as a therapeutic active ingredient.

25 16. A drug containing the crystalline form C, D or E of entacapone as claimed in claim 1, 2 or 3 and a therapeutically inert excipient.

17. The drug as claimed in claim 16 additionally containing levodopa and a decarboxylase inhibitor.

30 18. The use of the crystalline form C, D or E of entacapone as claimed in claim 1, 2 or 3, optionally in combination with levodopa and a decarboxylase inhibitor, for the treatment of Parkinson's disease or for the preparation of corresponding drugs.

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19. A process for the preparation of entacapone by a Knoevenagel condensation of 3,4-dihydroxy-5-nitrobenzaldehyde and N,N-diethyl-2-cyanoacetamide, charac-

terized in that the catalyst used for this condensation is diethylamine/acetic acid.

20. The process as claimed in claim 19, characterized
5 in that the N,N-diethyl-2-cyanoacetamide used has been prepared by reacting cyanoacetic acid with diethylamine in the presence of dicyclohexylcarbodiimide.

21. The process as claimed in claim 19 or 20,
10 characterized in that the 3,4-dihydroxy-5-nitrobenzaldehyde used has been prepared by the demethylation of 5-nitrovanillin with AlCl_3 /pyridine in chlorobenzene.